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REMARKS

Applicants thank Examiner Nguyen for the kind and courteous discussion held with Applicants' representative on February 17, 2004. During the discussion, Applicants' representative discussed the differences between silica gel and precipitated silica, and the outstanding rejections of the claimed invention in view of the cited references. The content of the discussion is reflected in the amendments filed concurrently herewith and the remarks contained herein.

Claim 1 is currently amended. Support for this amendment is found in original Claim 5. Claims 4-5 are cancelled without prejudice. No new matter will be added upon entry of the amendment. Upon entry of the amendment Claims 1-3 and 6-20 will be active, even though Claims 10-20 have been previously withdrawn from further consideration.

The rejection of Claims 1-4 and 6-9 under 35 U.S.C. § 103(a) as being unpatentable over Fujii et al. (U.S. 5,720,806) is obviated by amendment.

Fujii discloses "a filler for ink jet recording paper...which is composed of amorphous silica containing 0.3-1.0% by weight of aluminum oxide (Al_2O_3) and having a BET specific surface area of 250-400 m^2/g " (col. 2, ll. 42-46). Fujii does not suggest a composition of amorphous silica that contains an Al_2O_3 content of from 0.05 to 0.25% by weight and where the BET surface area is more than 300 m^2/g . In view of the fact that Fujii does not suggest reducing the Al_2O_3 content below 0.3% by weight, there can be no issue of obviousness. Therefore, it is requested that the Examiner withdraw this rejection.

The rejection of Claims 1-4 under 35 U.S.C. § 103(a) as being unpatentable over CN 1206020 (CN '020) is traversed. CN '020 discloses preparing silica gel. This is contrast to the present invention which claims precipitated silica. For the Examiner's convenience, Applicants enclose herewith a photocopy of a section from Kirk-Othmer Encyclopedia of Chemical Technology (vol. 20, Third Edition, pp. 768 and 773-777). The Examiner's

attention is directed to Table 1 and the accompanying pages, in which it is noted that silica gels and precipitated silica are recognized as being different materials. Therefore, in view of the fact that CN '020 discloses silica gel, it is requested that the Examiner withdraw this rejection.

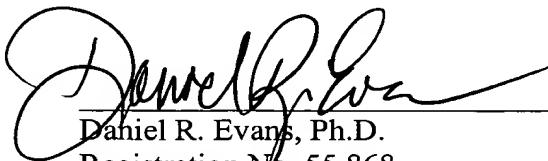
Finally it is requested that the provisional rejection of Claims 1-9 under the judicially created doctrine of obviousness-type double patenting as being unpatentable over Claims 1-20 of copending application No. 10/014,712, be held in abeyance until an indication of allowable subject matter.

It is requested that the Examiner acknowledge receipt of Applicants' priority document (DE 10048616.9, filed September 30, 2000), as Applicants filed a certified copy of said priority document on August 29, 2001. The Examiner's attention is directed to copies of Applicants' Request for Priority and face page of DE 10048616.9 enclosed herewith. These copies were retrieved from the USPTO's Private PAIR IFW system, and bear an August 29, 2001 date.

Finally, in view of the above-discussion, it is believed that Claim 1 is in a condition for allowance, which means that Claims 15-20 are allowable. It is requested that the Examiner rejoin these claims and pass the application to issue. An early and favorable indication of the same is respectfully requested.

Respectfully submitted,

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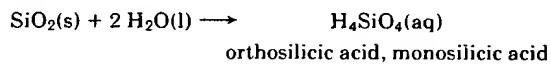
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Table 1. Properties of Different Forms of Amorphous Silica

| Property | Silica sols | Dry silica gels | Silica precipitated from solution | Pyrogenic silica |
|---|-------------|-----------------|-----------------------------------|------------------|
| SiO ₂ , % | 10-50 | 96.5-99.6 | 80-90 | 99.7-99.9 |
| CaO, % | na | na | 0.1-4 | na |
| Na ₂ O, % | 0.1-0.8 | 0-1 | 0-1.5 | na |
| wt loss, % | | | | |
| at 105°C | 50-80 | na | 5-7 | 0.5-2.5 |
| at 1200°C | 50-90 | 2-17.5 | 10-14 | 0.5-2.5 |
| ultimate particle size, nm | 5-100 | 1-100 | 10-25 | 1-100 |
| aggregate particle size, μm | | 3-25 | 1-10 | 2-3 |
| surface area, m ² /g | 50-700 | 200-700 | 45-700 | 15-400 |
| pH, aqueous suspension | 3-5, 8-11 | 2.3-7.4 | 4-9 | 3.5-8 |
| apparent or bulk density, g/cm ³ | 1.2-1.4 | 0.1-0.8 | 0.03-0.3 | 0.03-0.12 |
| true density, g/cm ³ | 2.2-2.3 | 2.22 | 2.0-2.1 | 2.16 |
| refractive index, <i>n</i> _D | 1.35-1.45 | 1.35-1.45 | 1.45 | 1.45 |
| oil absorption, g/g | | 0.9-3.15 | 1-3 | 0.5-2.8 |

Heterogeneous Reactions

Dissolution. Amorphous silica dissolves (or depolymerizes) in water according to the following equation:



$\text{H}_4\text{SiO}_4(\text{aq})$ can also be expressed as $\text{Si}(\text{OH})_4(\text{aq})$ or $\text{H}_2\text{SiO}_3(\text{aq})$. The solubility of amorphous silica in water at 25°C is in the range of 80-130 ppm SiO_2 (1.4-2.2 mmol/kg). This range of solubility is probably due to changes in the silica surface during dissolution. An area with a small positive radius of curvature dissolves most extensively, and an area with a small negative radius of curvature dissolves least extensively. The result is a flat surface of lower area and lower interfacial energy and hence lower solubility. The solubility of bulk silica at 25°C is ca 70 ppm SiO_2 (1.2 mmol/kg). Because of this surface chemistry, particle size affects solubility; particles with a diameter of less than ca 4 nm have a progressively greater solubility than larger particles (3). Several solubility studies have observed a decrease in solubility with time as the silica surface becomes more uniform, which makes determination of initial solubility difficult.

The solubility of amorphous silica in water increases with increasing temperature and increasing pressure (4). The solubility at 101.3 kPa (1 atm) and 0°C is ca 50 ppm SiO_2 (0.8 mmol/kg), increasing to 100 ppm (1.7 mmol/kg) at 25°C, and 750 ppm (12.5 mmol/kg) at 100°C. The solubility increases by ca 50% as pressure increases from 101.3 kPa to 101 MPa (1-1000 atm); the experiments were conducted in the 0-25°C range.

The solubility of amorphous silica in neutral dilute aqueous salt solutions is only slightly less than in pure water. The solubility in aqueous solutions at concentrations above ca 0.5 M decreases and is probably related to the amount of water available for hydration. Amorphous silica is essentially insoluble in methanol; solubility in methanol-water mixtures decreases with increasing percentage of methanol. Small amounts

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carbon), soluble salts, total solids, nonsiliceous ash, and metals (especially aluminum or iron). Physical characteristics include pH, density, viscosity, turbidity, refractive index, light-scattering properties, and sedimentation rate by ultracentrifugation. The particles are characterized by size and size distribution, porosity, degree of aggregation, specific surface area, and rate of dissolution. Particle size is measured by electron microscopy, light-scattering, absorbance of visible light, low-angle x-ray scattering, centrifugation, rate of reaction with molybdcic acid, or surface area measurement of dried particles (24) (see Analytical methods).

Modifications and Alterations. Coagulation, flocculation, or gelling and drying of silica sols produces amorphous silica powders. Gellation is rapid at pH 4-6. It is effected by increasing the silica content, eg, by evaporation. Further drying of the resulting gel yields colloidal aggregates. Coagulation of a sol is prevented by surface charge and surface hydration; conversely, neutralizing the surface charge by lowering the pH or adding salt causes coagulation. Addition of salts also causes surface adsorption of cations which reduce surface hydration. Polyvalent cations like Al^{3+} are strongly adsorbed by silica surfaces and neutralize the surface charge. Hydrated metal cations act as bridging compounds or flocculating agents for silica particles (see Flocculating agents). Flocculating agents used with silica sols are cationic surfactants such as octadecyltrimethylammonium bromide or organic polymers such as polyethyleneimine or polyacrylamide (21).

Sol particles are extremely small, and particulate silica obtained from sols is composed of aggregates or porous particles that have a much higher specific surface area than estimated from apparent size. Aggregate particles are also called secondary particles or clusters. Once silica particles have been formed as a sol, the surface can be modified by the attachment of different atoms or groups to obtain specific properties. Addition of aluminate ions gives an aluminosilicate surface that is much less soluble than the silica surface and much more stable toward gelling even at pH 4-6 and in the presence of salts. Addition of polyvalent metal oxides reverses the surface charge of silica and prevents bridging by siloxane bonds. Organic groups can be added to silica surfaces in the form of organic ions through formation of Si—O—C bonds as in esters or Si—C bonds as in organosilicon compounds. These surface alterations make silica easily dispersible in organic solvents including hydrocarbons. Some of these processes may cause the silica surface to become hydrophobic. Even a small change in particle surface characteristics may change the chemical behavior of the particle. Thus a silica particle covered with alumina behaves as an alumina particle, and a silica particle covered with a hydrocarbon coating acts as a large hydrocarbon molecule (25-29).

Silica Gel

Silica gels are classified into three types. Regular-density gel is made by gelling in an acid medium, which gives very small particles with high surface area (750-800 m^2/g). The average pore diameter is 2.2-2.6 nm, and the pore volume is 0.37-0.40 mL/g. Regular-density gel contains ca 6 wt % water as surface hydroxyl groups, which imparts a high capacity for water adsorption and adsorption of other polar molecules (see Drying agents). Regular-density gel exhibits a high selectivity for polar molecules and a large percentage of small pores. Intermediate-density silica has a lower surface area (300-350 m^2/g) but larger pore volume (0.9-1.1 mL/g). The average pore size is 12-16

nm in diameter, and the particles are larger than those of regular-density gel. Because of the large pore size, intermediate-density gel has a high capacity for water adsorption at high humidities. Intermediate-density gel is often used as a fine particle powder because particle size and porosity can be controlled. Low-density silica gel (eg, some aerogels) has a lower surface area (100–200 m²/g), larger average pore diameter (18–22 nm), and larger pore volume (1.4–2.0 mL/g) than the other types. It is usually prepared as a very fine powder of extremely low density; shrinkage of the gel during drying is minimized.

Properties. Silica gel is a coherent, rigid, continuous three-dimensional network of spherical particles of colloidal silica. A hydrogel is a gel in which the pores are filled with water, whereas a xerogel is a gel from which the liquid medium has been removed, causing the structure to collapse, thus decreasing porosity. If the liquid medium is removed in order to prevent shrinkage and collapse of the gel structure, an aerogel is formed. Porous glass is similar to a silica gel. Silica powder can be made by grinding or micronizing xerogels, which decreases the size of the gel fragments but leaves the ultimate gel structure unchanged. Gels and powders are characterized by the density, size, and shape of the particles, particle distribution, and by aggregate strength or coalescence (12–14,30–34).

When silica is used as an adsorbent, the pore structure determines the gel-adsorption capacity. Pores are characterized by specific surface area, specific pore volume (total volume of pores per gram of solid), average pore diameter, pore-size distribution, and the degree to which entrance to larger pores is restricted by small pores. These parameters are derived from gas- or vapor-adsorption isotherms, mercury-penetration studies, low-angle x-ray scattering, electron microscopy, gas permeability, ion or molecule exclusion studies, or measurement of volume of imbibed liquid (35).

Surfaces can be categorized as fully hydroxylated in which the surface consists solely of silanol (Si—O—H) groups, as in a siloxane (Si—O—Si), or as an organic surface. Silanol surfaces are formed by drying silica gels or precipitates from water below 150°C. These surfaces are readily wetted by water. Hydroxylated surfaces heated from 300–1000°C progressively develop a siloxane surface by dehydration; pyrogenic silicas also have siloxane surfaces. The behavior of particles with organic surfaces depends on the coating material. The particles may become dispersible in water, oil, or other organic solvents. If fluorocarbons are the surface group, the silica becomes both hydrophobic and oleophobic. The nature of the surface can be determined by measuring the heat of nitrogen adsorption, dye adsorption, infrared adsorption, or chemical analysis (14,18,26–28,36–37).

Preparation. Silica gels can be prepared by several methods (32,38). Most commonly, a sodium silicate solution is acidified to a pH less than 10 or 11; the gel time varies as shown in Figure 3.

The bulk-set process consists of the following steps: A silica hydrosol is prepared by mixing sodium silicate with a strong mineral acid. After it is allowed to set to a rigid mass, it is broken up mechanically. The silica concentration, temperature, and pH affect gelling time and final gel characteristics such as density, strength, hardness, surface area, and pore volume. The hydrogel particles are then washed free of electrolytes, and the washed gel is dried and activated. The rate of drying affects final gel properties.

In the slurry process, sodium silicate solution and acid are mixed, either batch or semicontinuously, to produce a gelatinous precipitate. This hydrogel is washed and

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consists in organic from water as heated pyrogenic surfaces water, oil, becomes refined by portion; or

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dried, often by spray-drying. Sodium silicate is the cheapest source of silica although natural sources can also be used. Certain clays yield relatively pure silica when leached with acid. Gels made from soluble silicates usually have very small particle size and the salts must be washed away before use. Gels can be made directly from salt-free colloidal silica, which provides a larger ultimate particle size and hence greater stability, along with low salt content. Surface characteristics may also be different, such as lower specific surface area and larger pore diameter.

Hydrolysis of pure silicon compounds like ethyl silicate, silicon tetrachloride, and other volatile hydrolyzable silicic esters is a third method of preparing gels. This method is more expensive, but produces very dense gels of high purity and very small pore size.

For some applications, silica gel is converted to pelletized or granular form by extruding pulverized gel with a binder or shaping the hydrogel during drying. Silica can be gelled in spherical form by spray-drying, or by spraying droplets into an immiscible liquid (emulsion polymerization). Freezing of a silica sol produces silica-gel particles of nonspherical shapes.

Characterization. The properties of a finished gel are determined by the size of the primary particles at the moment they aggregate into the gel network; the concentration of the primary particles in solution and thus the compactness of the gel network; the pH, salt concentration, temperature, and time during which the gel is aged while wet; mechanical pressure or shear forces applied to the gel before or during drying; temperature, pressure, pH, salt content, and surface tension of the liquid medium as it is being evaporated from the pores of the gel; and temperature, time, and type of atmosphere in which the gel is heated after being dried (39).

Modification. Once a gel structure is formed, it can be modified in the wet state to strengthen the structure or enlarge the pore size and reduce surface area (40). Gel reinforcement can be carried out in several ways: active or low molecular weight silica can be added to a broken-up gel in order to deposit it at a uniform rate, active silica can be added to a sol as the gel is growing which causes strong gel bridges to form between particles, or the wet gel can be heat-aged to increase coalescence of the particles. In this aging process silica is dissolved from smaller particles and deposited at the points of contact between larger particles, which causes strengthening. Washing can also be an aging step. Aging a wet gel increases interparticle bonding, which leads to less shrinkage of the gel during drying. The drying procedure affects the gel characteristics. Low density gels are made by minimizing shrinkage during drying by first reinforcing the gel. The wet gel is aged and the water is replaced with a liquid of lower surface tension, eg, alcohol. The gel is then heated to a temperature above the critical point of the liquid, thereby releasing the liquid as a vapor (aerogel process) (41).

Sintering a dried silica surface in air or in a vacuum causes shrinkage which decreases the surface area, whereas sintering in steam also increases pore size. Micropores are obtained by heating a hydrated gel at 1000°C for 10 h. The presence of impurities like aluminum tends to minimize changes caused by heating; however, at some temperature $>1000^{\circ}\text{C}$, and in the presence of impurities, silica gel is converted to cristobalite or to nonporous silica glass. Gels can be made with extremely small pores. Such gels include impervious silica, porous glass, and silica used as an adsorbent for certain specific materials which are determined by the surface composition and pore size of the silica gel.

Precipitated Silica

Precipitated silica (also called particulate silica) is composed of aggregates of ultimate particles of colloidal size that have not become linked in a massive gel network during the preparation process. Precipitated silicas are either formed from the vapor phase (fumed or pyrogenic silicas) or by precipitation from solution. Precipitated silica powders have a more open structure with higher pore volume than dried pulverized gels.

Silica can be precipitated from a sodium silicate solution using a lower concentration than in gel preparation. In the absence of a coagulant other than the sodium salt that is being formed, silica is precipitated from a hot sol at pH 9–10 when the concentration of sodium ion exceeds 0.3 N. Precipitation proceeds in several steps including nucleation of particles, growth of particles to desired size, coagulation to form aggregates by control of pH and sodium ion concentration, and reinforcement of the aggregate to the desired degree without further nucleation (42). Coagulating agents include sodium, calcium, or other polyvalent metal cations; ammonium ions; certain organic compounds; and fluoride ion. Reinforcement is carried out by adding active silica (silica that quickly dissociates to give monomeric silica) to the suspension of particles under alkaline conditions above 60°C. Silica is precipitated from silica sols by adjusting pH and salt concentration; care must be taken to prevent gelling (see Figs. 2 and 3). Silica is also precipitated by adding aqueous ammonium hydroxide to ethyl silicate, $(C_2H_5O)_4Si$, in alcohol.

Pyrogenic or fumed silica is prepared differently. Silica (usually sand) can be vaporized at ca 2000°C. On cooling, anhydrous amorphous silica particles form. In the presence of a reducing agent, eg, coke, silica sublimes at ca 1500°C to produce SiO , which can then be oxidized to produce particulate SiO_2 . Oxidation of silicon tetrachloride vapor at high temperature produces SiO_2 and Cl_2 . Alternatively, $SiCl_4$ can be burned with methane or H_2 to produce SiO_2 , H_2O , and HCl ; the latter process is an important commercial method. Silicon ester vapors can be oxidized and hydrolyzed to produce particulate silica of high purity though at high cost. SiF_4 , a by-product of the phosphate fertilizer industry, can also be used to produce silica by hydrolysis of the vapor at 1600–2200°C. HF is a product which can then react with sand to produce more SiF_4 (43–44).

The physical and chemical properties of precipitated silicas vary according to the manufacturing process. Ultimate and aggregate particle size in silicas precipitated from solution can be varied by reinforcement and control of suspension pH, temperature, and salt content. The particle size in pyrogenic silicas is controlled by combustion conditions. The surface area, as determined by nitrogen adsorption, is a function of particle size. Pyrogenic silicas tend to be less dense and more pure than silicas precipitated from solution since the latter contain coagulating agents. Pyrogenic silicas are much less hydrated and are sometimes completely anhydrous, whereas precipitated silicas may contain up to 10% water as surface hydroxyl groups that remain after drying at 150°C.

Some important chemical and physical properties of silica sols, silica gels, silica precipitated from aqueous solution, and pyrogenic silica are given in Table 1.

Naturally Occurring Amorphous Silica

Biogenic Silica. Several aquatic organisms, including diatoms, radiolarians, sponges, and silicoflagellates, secrete solid amorphous silica in the form of shells, skeletons, spines, or plates. These organisms extract silica from very dilute solutions (0.1 ppm SiO_2 , or 2 mmol/kg). These organisms are widespread, occurring both in marine and freshwater environments. They form a significant part of marine sediments in the equatorial Pacific (radiolarians) and in high latitude areas of all oceans (diatoms). However, most silica dissolves in water before becoming incorporated into the sediments. Dissolution is inhibited by incorporation of small amounts of metals and by the presence of an organic membrane coating over the silica (45) (see Diatomite).

Opal. Biogenic silica is sometimes called opal-a. With time opal-a becomes more structured, altering to opal-CT and then to opal-C, which is well crystallized cristobalite (46). These structural differences can be detected by x-ray diffraction or infrared spectroscopy. The gem opal is a cryptocrystalline (very fine-grained crystals) form of cristobalite with submicroscopic pores that contain water; the amount of water varies and can constitute several wt % (47).

Diatomaceous Earth. Diatomaceous earth [7631-86-9], also called kieselguhr or diatomite, is a loosely coherent chalk-like sediment made up of fragments and shells of diatoms (one-celled algae). It is used as an absorbent, filler, insulating material, and polishing agent. The particles are very fine and have high surface area; silica content may be as high as 94%. Initially, silica in diatomites was amorphous, but many of these deposits are millions (10^6) of years old, and the silica may now be present as cryptocrystalline quartz (47).

Chert. Chert is a diagenetic rock made up of microcrystalline or cryptocrystalline quartz with or without opaline silica. Most chert is white, tan, or gray and often contains other minerals. It may occur as nodules in limestone. Because of its hardness, chert is not very useful although it is sometimes substituted for sand in ceramics. Varieties of chert include flint, jasper, fossiliferous chert, oolite chert, novaculite, porcelainite, and tripoli. Most cherts have a biochemical origin and were originally composed of diatoms, radiolarian shells, or sponge spicules. These siliceous materials are transformed by dissolution, precipitation, recrystallization, and compaction over time to give a less soluble, denser material of lower surface area, which is called chert if granular, chalcedony if fibrous, or flint if dark gray. Some chert may originate from chemical precipitation. In these cherts, no traces of siliceous fossils remain (47).

Tripoli, a form of chert, is utilized as abrasive, filler, and extender. It is microcrystalline quartz ($<10 \mu\text{m}$ dia) although it is sold as amorphous silica (48). It is derived from calcite- or dolomite-bearing chert from which the carbonates have been leached, leaving a very pure silica. Tripoli is white or gray, soft, porous, and friable, and is mined extensively from Devonian deposits in Southern Illinois. Tripoli contaminated with iron oxide is red and has little commercial value (47) (see Abrasives; Fillers).

Amorphous Silica of Volcanic Origin. During the Roman Empire a colloidally subdivided high surface-area amorphous silica was mined at Pozzuoli, Italy, and on the Greek island of Santorini. This material is an alteration product of volcanic ash and when mixed with lime and sand gave an extremely impervious cement used to line cisterns (48).

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF: Juergen SCHUBERT, et al.

GAU:

SERIAL NO: NEW APPLICATION

EXAMINER:

FILED: HEREWITH

FOR: DOPED PRECIPITATED SILICA

J1036 U.S. PTO
09/940438
08/29/01


REQUEST FOR PRIORITY

#2

ASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, D.C. 20231

SIR:

Full benefit of the filing date of U.S. Application Serial Number, filed, is claimed pursuant to the provisions of 35 U.S.C. §120.

Full benefit of the filing date of U.S. Provisional Application Serial Number, filed, is claimed pursuant to the provisions of 35 U.S.C. §119(e).

Applicants claim any right to priority from any earlier filed applications to which they may be entitled pursuant to the provisions of 35 U.S.C. §119, as noted below.

In the matter of the above-identified application for patent, notice is hereby given that the applicants claim as priority:

| <u>COUNTRY</u> | <u>APPLICATION NUMBER</u> | <u>MONTH/DAY/YEAR</u> |
|----------------|---------------------------|-----------------------|
| GERMANY | 100 48 616.9 | September 30, 2000 |

Certified copies of the corresponding Convention Application(s)

are submitted herewith

will be submitted prior to payment of the Final Fee

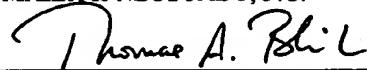
were filed in prior application Serial No. filed

were submitted to the International Bureau in PCT Application Number .
Receipt of the certified copies by the International Bureau in a timely manner under PCT Rule 17.1(a) has been acknowledged as evidenced by the attached PCT/IB/304.

(A) Application Serial No.(s) were filed in prior application Serial No. filed ; and
(B) Application Serial No.(s)
 are submitted herewith
 will be submitted prior to payment of the Final Fee

Respectfully Submitted,

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J1036 U S PTO
09/940438
08/29/01

Prioritätsbescheinigung über die Einreichung einer Patentanmeldung

Aktenzeichen: 100 48 616.9

Anmeldetag: 30. September 2000

Anmelder/Inhaber: Degussa AG, Düsseldorf/DE

Erstanmelder: DEGUSSA-HÜLS AKTIEN-
GESELLSCHAFT, Frankfurt am Main/DE

Bezeichnung: Dotierte Fällungskieselsäure

IPC: C 01 B, C 08 K, C 09 D

**Die angehefteten Stücke sind eine richtige und genaue Wiedergabe der ur-
sprünglichen Unterlagen dieser Patentanmeldung.**

München, den 28. Mai 2001
Deutsches Patent- und Markenamt
Der Präsident
Im Auftrag

Wobinger